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EXPERIMENTAL STUDIES ON SUPERCRITICAL FLOWS

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19980921 023

DTIC QUALITY INSPECTED 1

AFRL-SR-BL-TR-98-

0623

REPORT DOCUMENTATION PAGE

1. AGENCY USE ONLY (Leave blank)	2. REPORT DATE 20 August 1998	3. REPORT TYPE AND DATES COVERED Annual Technical Report

4. TITLE AND SUBTITLE	5. FUNDING NUMBERS
Experimental Studies on Supercritical Flows	PE - 61102F PR - 2308 SA - BS C - F49620-97-C-0002
6. AUTHOR(S) Gregory W. Faris	

7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) SRI International 333 Ravenswood Avenue Menlo Park, CA 94025	8. PERFORMING ORGANIZATION REPORT NUMBER MP 98-065

9. SPONSORING / MONITORING AGENCY NAME(S) AND ADDRESS(ES) AFOSR/NA 110 Duncan Avenue, Suite B115 Bolling AFB DC 20332-0001	10. SPONSORING / MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

12a. DISTRIBUTION / AVAILABILITY STATEMENT Approved for public release; distribution is unlimited	12b. DISTRIBUTION CODE

13. ABSTRACT (Maximum 200 Words)

Experimental apparatus was assembled for performing stimulated scattering. An initial setup used the fundamental radiation from an injection-seeded Nd:YAG laser as a pump laser, and a tunable laser diode as a probe laser. However, the two tunable laser diodes provided by the manufacturer stopped working prematurely, and the manufacturer has not been able to provide us with a working laser. Thus, a second experiment was set up using the second harmonic radiation from the Nd:YAG laser as a pump laser and an argon-pumped ring dye laser as a probe laser. With this arrangement stimulated Brillouin scattering was obtained in glasses and methanol. Specific system improvements were made to improve signal-to-noise ratios including bandpass electrical filtering, single-shot measurements, and triggering on the laser light. With these improvements, stimulated Rayleigh scattering in methanol was observed. These experiments provided information on the requirements of a cell for supercritical experiments.

14. SUBJECT TERMS Supercritical fluids, stimulated scattering, Raman scattering, Brillouin scattering, Rayleigh scattering.			15. NUMBER OF PAGES 16
			16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT Unclassified	18. SECURITY CLASSIFICATION OF THIS PAGE Unclassified	19. SECURITY CLASSIFICATION OF ABSTRACT Unclassified	20. LIMITATION OF ABSTRACT UL

NSN 7540-01-280-5500

Standard Form 298 (Rev. 2-89)
Prescribed by ANSI Std. Z39-18
298-102

OBJECTIVES

The objectives of this research are to develop stimulated scattering as a diagnostic for supercritical fluids, and use this technique to improve our understanding of fluids in the supercritical state.

The study of supercritical fluids and flows requires new diagnostic techniques. Currently available techniques such as laser-induced fluorescence (LIF) and coherent anti-Stokes Raman scattering (CARS) are complicated by increased molecular interactions, leading to stronger quenching, larger absorption and refractive index, and incomplete understanding of the influence of local conditions on spectroscopic parameters such as linewidths, nonresonant background contributions, and quenching rates. We believe that stimulated scattering techniques hold great promise for studying supercritical fluids.

STATUS OF EFFORT

Experimental apparatus was assembled for performing stimulated scattering. An initial setup used the fundamental radiation from an injection-seeded Nd:YAG laser as a pump laser, and a tunable laser diode as a probe laser. However, the two tunable laser diodes provided by the manufacturer stopped working prematurely, and the manufacturer has not been able to provide us with a working laser. Thus, a second experiment was set up using the second harmonic radiation from the Nd:YAG laser as a pump laser and an argon-pumped ring dye laser as a probe laser. With this arrangement stimulated Brillouin scattering was obtained in glasses and methanol. Specific system improvements were made to improve signal-to-noise ratios including bandpass electrical filtering, single-shot measurements, and triggering on the laser light. With these improvements, stimulated Rayleigh scattering in methanol was observed. These experiments provided information on the requirements of a cell for supercritical experiments.

ACCOMPLISHMENTS/NEW FINDINGS

We are using stimulated scattering (stimulated Rayleigh, Brillouin, and Raman scattering) to study supercritical fluids. New diagnostics are needed in the supercritical regime because low pressure diagnostics do not work well. From our measurements we can determine thermal, compressional, and compositional properties of supercritical fuels. These techniques should improve our knowledge of fluid properties in the supercritical state and allow *in-situ* diagnostics of supercritical fuels.

STIMULATED SCATTERING

Rayleigh, Brillouin, and Raman scattering occur commonly as spontaneous scattering. These scattering processes arise from natural oscillation modes of materials and can be used to determine the physical parameters responsible for those oscillations. When these collective modes are excited with a powerful laser, the mode oscillations can be driven so hard that they grow exponentially. In this case, the oscillations cause stimulated scattering. The dominant advantage of stimulated scattering is that the scattered signal can be made arbitrarily large; otherwise, these processes produce extremely weak signals. By using a probe to measure the induced amplification, we can obtain very good quantitative results. This technique is distinct from the stimulated scattering that builds up from noise, in which case quantification is very difficult.

The large signals from stimulated scattering are particularly helpful for investigating Rayleigh and Brillouin scattering, where the weak signals available from spontaneous scattering are difficult to discriminate from background excitation light. Other advantages of stimulated scattering include exceptional temporal resolution, and improved spectral resolution and signal-to-noise ratio. Furthermore, the use of two laser beams allows spatial registration and point measurement of local conditions.

With a single detection system, all three processes—Rayleigh, Brillouin, and Raman—can be measured. These processes together provide measurements of a wide range of material properties. Rayleigh scattering provides information on thermal properties, Brillouin scattering on compressional or elastic properties, and Raman scattering on chemical and compositional properties, density, and temperature. While spontaneous Brillouin[1] and Raman[2,3,4] scattering have been applied to supercritical fluids, the use of stimulated scattering for supercritical fluids is new.

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EXPERIMENT

The first phase of our work is directed primarily at measurements of the properties of bulk supercritical materials in static or flowing cells. This work will allow measurements of the physical properties of supercritical fuels, including compressibility, speed of sound, thermal diffusivity, and chemical composition, density, and temperature. This phase is significant, because current work on supercritical fuels is limited to measurements performed after returning the fuels to ambient conditions, rather than in the supercritical state. With experience gained from measurements on uniform supercritical fluids, we expect that we will then be able to apply these techniques to supercritical mixing and combustion.

Stimulated scattering measurements are performed by producing strong scattering interactions using a pump laser and then probing the scattering using a second probe laser. The experimental arrangement for this type of measurement is shown in Figure 1. The pump laser sets up an electric polarization oscillating at the characteristic frequency of a scattering mode of the material.[5] For strong laser driving, this polarization acts as a driving force, leading to amplification of both the material oscillation and a scattered optical wave. The optical amplification is detected as a gain or loss on the probe beam. Spatial resolution is determined by the overlap volume of the pump and probe beams.
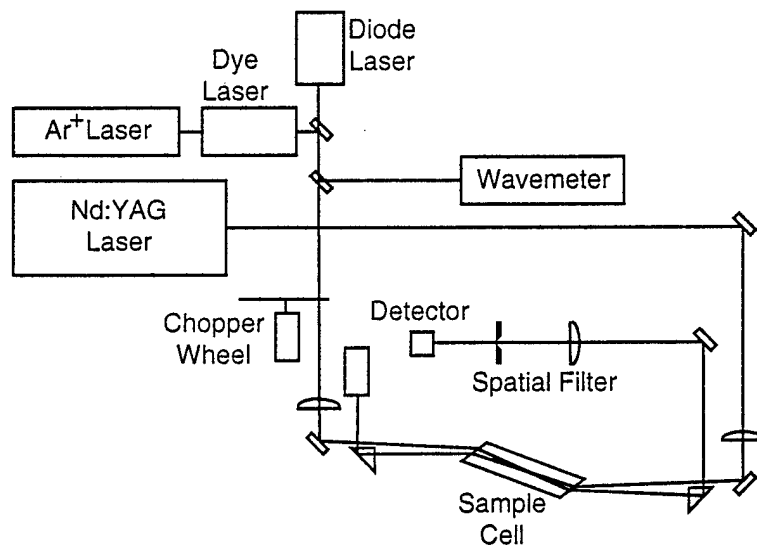

Figure 1. Experimental arrangement for stimulate scattering measurements.

As a pump laser for our stimulated scattering measurements we use an injection-seeded Nd:YAG laser. This laser has been modified previously allowing production of linewidths down to 20 MHz and below.[5] This narrow linewidth is possible through generation of long pulse lengths produced through lowered oscillator gain and pulse lengthening in the amplifier. Narrow

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linewidths for the pump laser are important for resolving Brillouin features which can be as narrow as 13 MHz.[6] The injection-seeded Nd:YAG laser has not been operated in a narrowband for several years, and required replacement of several optics and complete realignment. The laser is now operating very well.

Because stimulated scattering measurements are nonresonant, they can be performed at any wavelength for which the medium is transparent. Much of our previous measurements were performed at 532 nm.[7,8] The pump beam was the second harmonic of an injection-seeded Nd:YAG laser. The probe beam was produced with a cw dye laser pumped by an argon ion laser. Recent advances in diode laser technology allow use of a tunable diode laser as the probe laser. In this case, the fundamental of the Nd:YAG laser is used as the pump laser and measurements are performed at 1.064 μm.

The wavelengths of 532 nm and 1.064 μm each have their relative advantages. The fundamental wavelength has lower absorption for some of the samples we wish to study, such as thermally stressed jet fuel.[9] While this capability is desirable, it is not essential to our studies. Use of a diode laser probe beam also avoids the large operating costs for the argon laser and reduces the amount of time required for keeping the argon and dye lasers in good operation. On the other hand, operation at the second harmonic provides larger gain coefficients for stimulated scattering. For example, for Brillouin scattering, the gain coefficient should be about four times larger at 532 nm than at 1.064 μm.

We plan to perform to perform most of our measurements at 1.06 μm, using the Nd:YAG laser fundamental as the pump radiation and a diode laser as the probe laser. We originally proposed to modify a high power tunable diode laser from SDL for these measurements. This laser can provide up to 0.5 W, but is not narrow band. Given the amount of effort required to convert this laser to narrowband operation, we have chosen to use a lower power diode laser from Environmental Optical Sensors, Inc. that already operates narrowband. Although there are potential complications involved with scattered pump light when using a lower power probe laser, these are expected to be less involved than converting the high power laser to narrowband operation.

Scattered pump light can appear to be a spurious gain signal on the probe laser. Significant discrimination against the scattered pump beam is achieved by spatial filtering of the probe beam prior to detection. However, as the power of the diode laser is low, other means of reducing scattered pump light may be necessary. Cell design can reduce this scattered light as described below. Another approach is to amplify the diode laser using either an anti-reflection coated laser diode or a fiber amplifier similar to the Yb-doped fiber lasers which have recently

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become available. A third approach is to use frequency modulation (fm) techniques to discriminate against the pump light. Our diode laser has a bias tee to allow use of high frequency modulation for fm detection.

A low pressure sample cell has been constructed that has a length longer than the overlap between the pump and probe lasers. This removes the scattered pump light from the line of sight along the probe beam. For a given probe power, the scattered pump light places a limit on the minimum size of the stimulated gain signal. The cell has Brewster angle windows to further reduce scattered light.

Environmental Optical Sensors Inc. (EOSI) produces a tunable diode laser with single mode operation tunable over 30 to 40 nm around 1.064 μm with close to 10 mW of power. A laser was purchased from EOSI to perform our measurements. Although EOSI had supplied lasers at this wavelength in the past, they have had a number of difficulties in supplying our laser. Shipment of the laser was delayed due to difficulties in applying the anti-reflection coating required for single mode operation of this laser. When the laser was shipped, it died within the first two weeks. A second laser was sent, which also died prematurely. The EOSI has supplied three lasers at this wavelength to other researchers this year, so we are confident that they can ultimately provide us with a laser. However, as of this date we do not have a functioning laser.

The rest of the experimental apparatus for 1.064 μm stimulated scattering measurements is in place, including the pump laser, detection electronics, and full integration of the tunable diode laser driver to a computer data acquisition package. The diode laser module has been interfaced to a PC-compatible computer to allow automated measurement of stimulated scattering spectra. Before the first laser died, we tested the laser amplitude noise and found it to be within an acceptable range for our measurements. Excessive laser amplitude noise would prevent measurements of small gain signals. We are ready to take data for the 1.064 scattering measurements as soon as the laser arrives.

We have demonstrated that the cell and pump laser are easily capable of generating stimulated scattering signals by producing stimulated Brillouin scattering from dichloroethane without a probe beam. In this case, the stimulated scattering builds up from a single photon spontaneously scattered by the sample. This requires gains of exp(30) or more, or much more than that required for our stimulated scattering measurements.

Because we do not currently have an operating laser diode for the probe laser, we have set up additional lasers to operate at 532 nm. We have borrowed an Ar+ laser and cw ring dye laser for this purpose. Switching between 532 nm and 1.064μm operation is performed by inserting or removing the doubling crystal in the Nd:YAG laser.

Using the 532-nm pump and probe lasers, we have obtained stimulated Brillouin and stimulated Rayleigh signals in subcritical materials. Stimulated Brillouin scattering has been obtained both in glasses and in methanol. A spectrum for stimulated Brillouin scattering in methanol is shown in Figure 2. There are two peaks in the spectrum. The positive-going peak at the left is a gain peak, which occurs when energy is transferred from the pump laser to the probe (dye) laser. The negative-going peak at the right is a loss peak, which occurs when energy is transferred from the probe laser to the pump laser. Information that can be extracted from these peaks include the overall density (from the peak height), the acoustic velocity or compressibility (from the peak separation) and the acoustic lifetime or phenomenological viscosity parameter (from the peak widths).
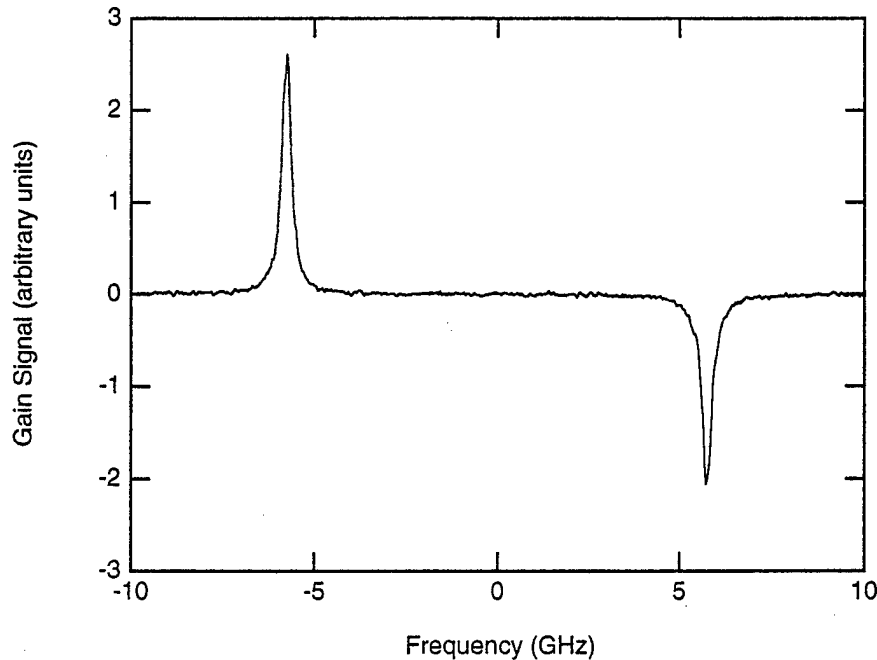

Figure 2. Stimulated Brillouin scattering spectrum for methanol.

A number of experimental improvements were required in order to obtain a stimulated Rayleigh scattering signal. The dye laser has large intensity fluctuations both near 1 MHz and near 80 MHz. These were reduced by using low and high pass filters that transmitted most of the frequency components in the pump pulse. Amplitude fluctuations associated with jitter of the pump pulse were reduced by triggering the boxcar used for data acquisition from the laser light rather than the Q-switch trigger pulse. The data acquisition electronics were configured to perform single shot measurements. This allows subtraction of the scattered pump laser light on a

shot-by-shot basis. Following the signal-to-noise improvements, we have been able to observe stimulated Rayleigh scattering in methanol. This peak is shown in Figure 3 along with a fit to the appropriate theoretical lineshape.
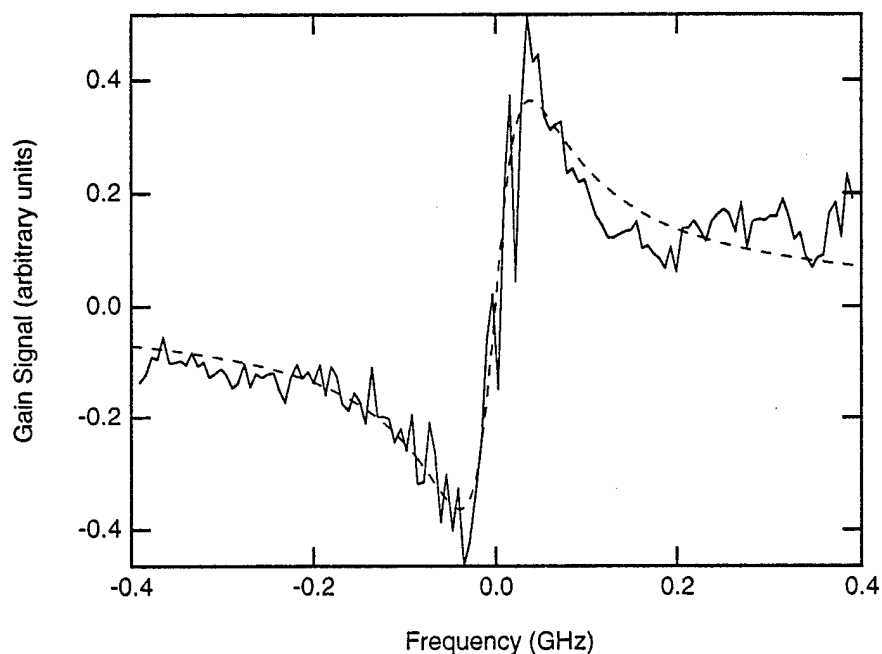

Figure 3. Stimulated Rayleigh spectrum for methanol.

Observation of stimulated Brillouin and stimulated Rayleigh scattering in methanol at low pressures helps define the design requirements for a high pressure cell that will allow measurements in the supercritical regime. A longer cell allows reduced beam overlap on the windows, which in turn reduces the scattered light background signal. However, the longer cell can also increase the production of stimulated Brillouin scattering in absence of the seed laser, which also causes background signals. We find that careful alignment can prevent stimulated Brillouin scattering in a long cell, making it the better design. Our experiments have confirmed that we will need tilted windows for the high pressure cell. The cell design in ongoing, and construction will begin soon.

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9. G. W. Faris, "Experimental Studies on Supercritical Flows," in ARO/AFOSR Contractors Meeting in Chemical Propulsion, Ohio Aerospace Institute, June 17-19 1997, pp. 112-115.

PERSONNEL

The following professional scientists participated in the research supported by this contract:

Gregory W. Faris, Research Physicist, principal investigator and lead experimentalist.

Yihong Chen, Postdoctoral Fellow, experimentalist.

DaNel Hogan, Undergraduate Student, experimentalist.

PUBLICATIONS

The following publications were made on research supported by this contract or the previous contract:

S. A. Meyer and G. W. Faris, "High Power Lyman Alpha Source Generated with an ArF Excimer Laser," Opt. Lett. **23**, 204-206 (1998).

W. Juchmann, J. Luque, and J. B. Jeffries, "Atomic Hydrogen Concentration in a Diamond Depositing dc-Arcjet Determined by Calorimetry," J. Appl. Phys. **81**, 8052 (1997).

W. Juchmann, J. Luque, J. Wolfrum, and J. B. Jeffries, "Absolute Concentration, Temperature, and Velocity Measurements in a Diamond Depositing dc-Arcjet Reactor," Diamond and Diamond Related Mat., **7**, 165 (1998).

J. Luque, W. Juchmann, E. A. Brinkman, and J. B. Jeffries, "Excited State Density Distributions of H, C, C2, and CH by Spatially Resolved Optical Emission in a Diamond Depositing dc-Arcjet Reactor," J. Vacuum Sci. and Tech. A., **16** 397 (1998).

The first publication is attached as the appendix. The remaining three publications have been sent under a separate cover letter.

INTERACTIONS/TRANSITIONS

PRESENTATIONS

The following presentations were made on research supported by this contract or the previous contract:

Scott A. Meyer and Gregory W. Faris, "Phase Matching Effects on VUV Generation near Lyman Alpha," Paper MGG2 presented at the Optical Society of America Annual Meeting, 13th Interdisciplinary Laser Science Conference, Long Beach, California, 12-17 October 1997.

G. W. Faris, "Experimental Studies on Supercritical Fluids," presented at the ARO/AFOSR Contractor's Meeting in Chemical Propulsion, Long Beach, California, 29 June - 1 July 1998.

INTERACTIONS

The following interactions occurred at the AFOSR/ARO contractors meeting in Chemical Propulsion, Long Beach, California, 29 June - 1 July 1998.

Tim Edwards, supercritical fuels, diagnostic requirements, and supercritical cells.

Mel Roquemore, miscellaneous research on combustion and fuels.

Tom Jackson, high pressure and hypersonic diagnostics.

Irv Glassman, supercritical flow cells, carbon structures

Yaw Yeboah, supercritical cells and diagnostics

Pablo Debennedetti, supercritical mixtures

Doug Talley, supercritical diagnostics

Jerry Seitzman, laser diagnostics,

Michael Winter, diagnostics and sensors

Med Colket, combustion chemistry

Peter DeBarber, diagnostics

Bob Santoro, absorption spectra of organics

Ron Hanson, diagnostics

Marshal Long, tomography

Ian Kennedy, diagnostics,

Nina Bergan French (Sky Plus)and Steve Priebe (INEL), laser diagnostics, environmental monitoring.

TRANSITIONS

Performer: Dr. G. W. Faris, SRI International, 650 859 4131

Customer: Dr. Surendra Sharma, NASA Ames Research Center, 650 604 3432

Result: VUV generation using Raman-shifted ArF laser

Application: Measurement of oxygen concentrations for electric arc shock tube experiments for high speed aerodynamics research and development.

INVENTIONS

None.

HONORS/AWARDS

Gregory Faris, ARCS Fellow, 1985-1986

APPENDIX

HIGH POWER LYMAN ALPHA SOURCE GENERATED WITH AN ARF EXCIMER LASER, OPT. LETT. 23, 204-206 (1998).

High-power Lyman-α source generated with an ArF excimer laser

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We report high-power vacuum-ultraviolet (vuv) generation at the Lyman-α wavelength of 121.6 nm, using a simple experimental system. vuv radiation is produced through two-photon-resonant difference-frequency mixing with a tunable ArF excimer laser and a Nd:YAG-pumped dye laser. Using phase-matched mixtures of Kr and Ar at a total pressure of 650 mbar, we produced 7-μJ energies at Lyman-α in approximately 5 ns (1.3 kW), as measured directly with a pyroelectric energy probe. Measurements indicate that higher powers are possible with system optimization. A tuning range of 0.1 nm was achieved for a fixed gas mole fraction at a total pressure of 650 mbar. Qualitative agreement is found between measured tuning profiles and theoretical predictions. © 1998 Optical Society of America
    OCIS codes: 190.0190, 140.7240, 140.2180.

Development of coherent vacuum-ultraviolet (vuv) sources continues to provide new opportunities for vuv spectroscopy and photochemistry. Higher powers and broad tunability are extending techniques such as multiphoton detection, planar imaging, and photolysis deeper into the vuv. Hydrogen Lyman-α radiation (121.567 nm) is especially interesting because it permits single-photon detection of ground-state H atoms, which are important in arc-jet plumes, tokamaks, combustion, molecular dynamics, and astrophysics. Several researchers have generated Lyman-α with dye lasers and phase-matched mixtures of Kr with Xe or Ar by use of frequency tripling[1-3] or difference-frequency mixing.[4]

Alternatively, substantial vuv power can be generated with an ArF excimer laser and a dye laser by two-photon-resonant difference-frequency mixing, $\nu_{vuv} = 2\nu_{ArF} - \nu_{dye}$.[5-7] One can enhance this process by tuning the ArF laser to a two-photon resonance in H_2, HD, or Kr, and tuning is provided by variation of the wavelength of the dye laser. By varying the dye-laser wavelength, one can produce vuv wavelengths of 110 to 180 nm. Because of the importance of the Lyman-α wavelength, our objective was to evaluate the energies available at Lyman-α with this technique. Kr has negative dispersion near Lyman-α, allowing phase matching by use of gas mixtures.

A schematic of the experimental arrangement is shown in Fig. 1. Because of the importance of beam quality for nonlinear frequency conversion, we modified the ArF laser (Lambda Physik EMG-150 MSC) to provide spatial filtering of the oscillator output beam and triple pass the amplifier.[8] This laser is tuned to two-photon resonances in the mixing gas. The third photon is provided by a Nd:YAG-pumped dye laser (Quanta-Ray DCR II and PDL) using Lambda Physik LC4700 dye. Output energies of the ArF laser and the dye laser are typically 20 and 10 mJ, respectively, and linewidths are of the order of 1 cm^{-1}. The beams are combined on a high-power ArF dielectric mirror and focused with a 50-cm lens into the gas cell. We verify spatial overlap by temporarily directing portions of the beams through a pinhole by use of a wedge. Temporal overlap is maintained by a drift-control circuit and verified by a fast photodiode. To minimize optics transmission losses, we use a single MgF$_2$ plano-convex lens (50-cm focal length at 250 nm) to provide an exit window for the mixing cell, collimate the vuv beam, and dispersively separate the vuv beam from the ArF and the dye beams. An aperture of 7-mm diameter spatially selects the vuv beam, which is detected by a Laser Precision RjP735 pyroelectric energy meter and a KBr photomultiplier tube. Two new vuv-grade MgF$_2$ lenses purchased from Optovac and Harshaw–Bicron were used for these measurements, and both initially yielded >70% transmission at Lyman-α. To date we have used only the Optovac lens and have not encountered the same degradation that was encountered in the research reported in Ref. 9, perhaps owing to improvements in MgF$_2$ growth techniques.

Although the pyroelectric energy probe allows relatively straightforward measurement of the vuv energies, it is not without complications. Irradiation of the probe with high-energy vuv radiation near Lyman-α causes the energy-probe output to decay within a period of minutes, becoming less sensitive to the ArF and the dye beams as well as to the vuv beam. The sensitivity recovers over periods of 10 min to several hours. The cause of this detector-sensitivity drop is
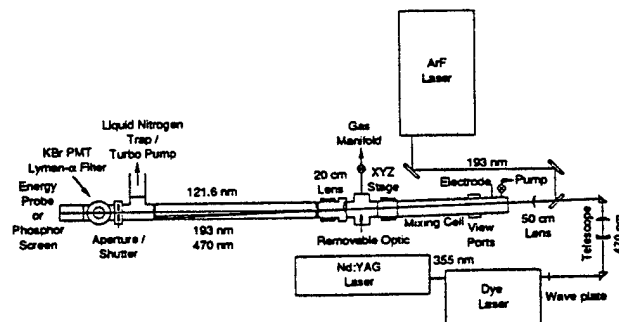

Fig. 1. Experimental apparatus for vuv generation. PMT, photomultiplier tube.

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not currently understood. To provide a stable energy monitor, we also use a solar-blind KBr photomultiplier with a Lyman-α filter to measure vuv light scattered from the probe at right angles to the beam. This photomultiplier–filter combination is insensitive to ArF and dye light, and signal-to-background ratios of up to 100 are obtained. We estimate the accuracy of our energy measurements at 10% for direct probe readings and 30% with the photomultiplier and the filter.

Without phase matching, mixing in H_2 produces 2 orders of magnitude more energy than mixing in Kr near Lyman-α. However, H_2 does not produce vuv radiation well at Lyman-α because of production of H atoms, which absorb the vuv. In addition, resonant contributions to the refractive index from H atoms produce large variations in phase mismatch, causing large intensity fluctuations with varying vuv wavelength. Through manipulation of experimental parameters such as H_2 pressure, laser powers, laser pulse synchronization, and ArF wavelength, we were not able to reduce H atom production sufficiently to allow good conversion at Lyman-α. For this reason we investigated the use of Kr.

One can enhance difference-frequency mixing in Kr by tuning the ArF laser to two-photon resonances with the $6p[5/2,2]$, $6p[3/2,2]$, and $6p[1/2,0]$ states. Transitions to $6p[5/2,2]$ and $6p[1/2,0]$ lie in the wings of the ArF tuning curve, so we chose the $6p[3/2,2]$ transition. We purged the ArF beam path with argon to minimize losses owing to oxygen Schumann–Runge absorptions at this wavelength.[8] Generation of vuv radiation in pure Kr is poor because of the relatively large phase mismatch in Kr. The pressure dependence of the vuv intensity in pure Kr is shown in the lower part of Fig. 2. Peak energies of ~0.2 μJ are produced at an optimum pressure of 7 ± 3 mbar, roughly an order of magnitude lower than the optimum pressure in H_2. This is consistent with a calculated phase mismatch for Kr, which is five times larger than for H_2 at Lyman-α. Phase mismatch can be eliminated by the addition of a positively dispersive gas such as Ar but not H_2, because H atoms are produced even when the ArF laser is tuned to the Kr resonance.

With phase-matched mixtures, higher partial pressures of Kr can be used. Figure 2 shows that a 3.9:1 Ar/Kr mixture with 260 mbar of Kr yields roughly 2 orders of magnitude of improvement over pure Kr. Phase-matched vuv generated at 130 mbar of Kr is shown in Fig. 3. We measured as much as 7 μJ of energy with 130 mbar of Kr. The excimer laser has a pulse length much longer than that of the dye laser, so we use the dye pulse length (5 ns FWHM) as an estimate of the vuv pulse length. At this pulse length we produced 1.3 kW of power at Lyman-α. As an indication of the brightness of our source, the Lyman-α radiation can be observed on a phosphor screen with the room lights on. The highest vuv power at Lyman-α, that was previously reported apparently corresponds to 250 W of power external to the mixing cell.[4] This previous power was measured indirectly through comparison of dye and vuv intensities measured by a spectrometer and a photomultiplier. This measurement approach is inaccurate owing to a variety

of systematic errors, such as comparing beam powers based on the fraction of the beams sampled by the spectrometer slit.

Our experiments indicate that significantly higher vuv powers are possible with further system optimization. Figure 2 shows that the vuv power is still increasing with pressure. Our cell was not designed for operation at overpressure, so higher total pressures were not explored. Further improvement is also possible by using larger focal-spot sizes (longer-focal-length lenses). We did not optimize the focal length of the focusing lens. Because the generated vuv depends on the path-integrated laser intensities, it varies with the incident laser powers, not with the peak intensities in the foci.[10,11] Thus larger focal-spot sizes can reduce saturation effects (which increase with the peak intensity) without sacrificing vuv power. Indications of saturation, which can occur as a result of dielectric breakdown, multiphoton ionization, ground-state depletion, intensity-dependent refractive-index changes, or other processes, are present in our measurements. Ideally, the vuv power scales with the square of the density, and our power rises more slowly (see Fig. 2). To gain full advantage of the longer conversion path lengths obtained with larger focal-spot sizes, one must
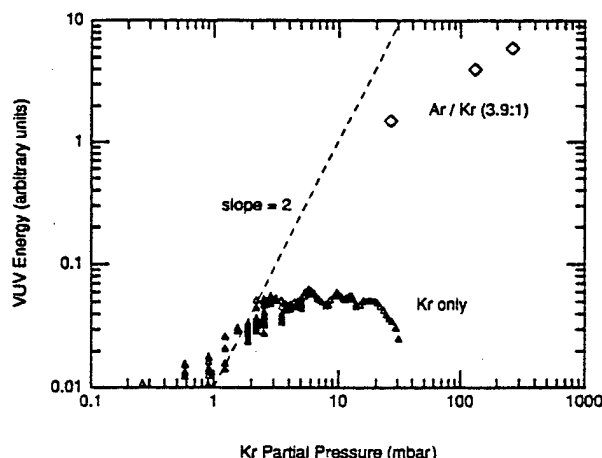

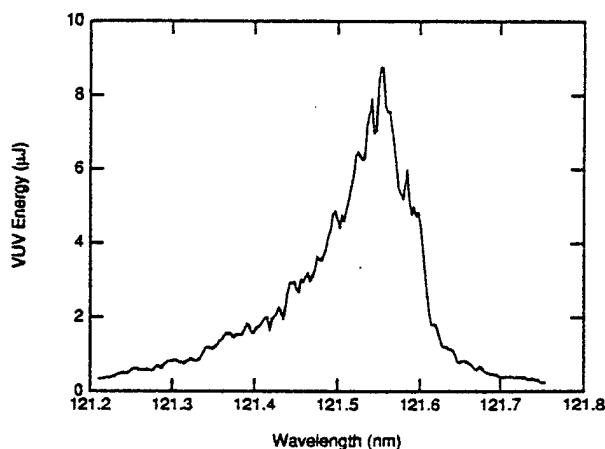Fig. 2. Generated vuv as a function of Kr partial pressure. All measurements were performed at Lyman-α.

Fig. 3. Absolute vuv energy for a Kr partial pressure of 130 mbar. 7 μJ is generated at Lyman-α.
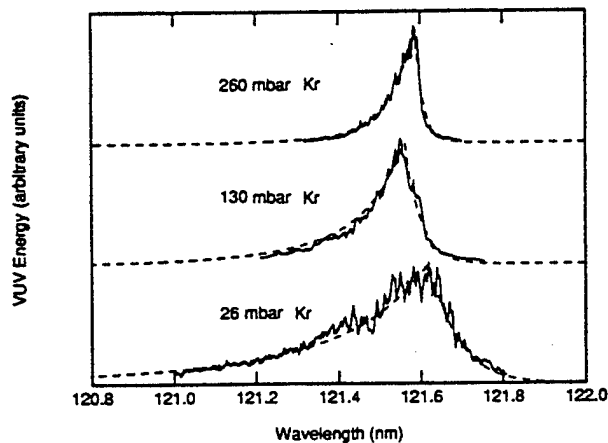
Fig. 4. Tuning range of vuv at different partial pressures of Kr. Solid curves, experimentally measured tuning profiles; dashed curves, fits using theoretical profiles.

ensure that the beam quality is quite good, which may require phase-conjugation amplification of the ArF laser.[12] Breakdown is often observed in the focal region at higher total pressures. Some reduction in breakdown or other saturation processes may be possible by use of a buffer gas with a higher refractive index than Ar, as the mixture used here is only 20% Kr. For operation at high pressures, absorption and (or) dispersion by rare-gas dimers may become important.[1] Because the tuning range at a fixed gas mixture is inversely proportional to the laser confocal parameters and the gas pressure, increased power may come at the cost of reduced tuning range. However, the tuning range may be increased by using a gas mixture that has a flatter dispersion than Ar/Kr.

The spectral tuning ranges for three Kr partial pressures are shown as solid curves in Fig. 4. As expected, the tuning range decreases (approximately) linearly with increasing pressure. These tuning profiles show a strong asymmetry, too strong to be caused by a curvature in the phase mismatch as a function of wavelength. Such asymmetry can be caused when ArF and the dye beams have dissimilar confocal parameters. This asymmetry is related to the additional phase change that occurs when the beams pass through the focus (Guoy effect). When the confocal parameters are not equal, a ripple is produced on the phase of the driving polarization that generates the vuv radiation. Depending on whether the phase of the driving polarization leads or lags the phase of the vuv wave (i.e., whether the phase mismatch is negative or positive), the phase ripple can be compensated for to a greater or lesser extent, leading to an asymmetry.

We fitted phase-matching curves to the profiles in Fig. 4 based on the expressions in Ref. 11.[13] These fits are shown as dashed curves in Fig. 4. The fitted confocal parameters for the ArF beam are 1.6 mm for 26 mbar of Kr and 6 mm for the two higher pressures. The fitted confocal parameters for the dye

laser are four times smaller than the ArF confocal parameter in each case. If we use the low-pressure confocal parameters to calculate focal-spot sizes, we find that the calculated and the measured spot sizes agree for the ArF beam, whereas the calculated spot size is roughly half the measured spot size for the dye laser. Our lasers do not have diffraction-limited Gaussian beams, so some disagreement in these values is not surprising. The fact that the fitted confocal parameters are not the same for all pressures may be related to refractive-index changes induced by the lasers or to other saturation effects. The dependence of the asymmetry on experimental confocal parameters means that the direction and the amount of asymmetry are experimentally dependent. The vuv beam profile also depends on the phase mismatch Δk and so will vary with wavelength.[10]

We have achieved directly measured energies of 7 μJ at Lyman-α, using two-photon-resonant difference-frequency mixing in phase-matched gas mixtures of Kr and Ar. Significant improvement in energy is expected with further system improvement, although this may come at the expense of a reduced tuning range for a fixed gas mixture.

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